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Relaxation oscillations in long-pulsed random lasers

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We have measured the evolution of the light intensity of a random laser during a nanosecond pump pulse. Relaxation oscillations in a titania random laser were observed in the time trace of the total emitted intensity. We compare our experimental results with a simple model, based on the four-level rate equations for a single mode laser.

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I. INTRODUCTION

Relaxation oscillations of conventional lasers are a well-understood phenomenon.[1] They are especially important for continuous wave and long-pulsed lasers. In a random laser, a medium in which gain is combined with multiple scattering of light, relaxation oscillations can also occur, as was predicted by Letokhov in 1968.[2] A pre-eminent experimental demonstration of a random laser was published by Lawandy *et al.* in 1994 [3], followed by many others [4, 5, 6, 7]. Only recently Soukoulis and coauthors have presented measurements of relaxation oscillations in single modes of a picosecond pumped random laser system.[8] Different numerical calculations on random lasers also show this oscillatory behavior.[9, 10, 11] To our knowledge, no measurements have been performed on relaxation oscillations in random lasers in the interesting regime of long pulses.

In this report we present measurements of the time evolution of a nanosecond pumped random laser system. We compare our experimental observations with a simple model, based on the four-level rate equations for a single-mode laser.

II. A SIMPLE MODEL

The evolution of the excited molecules N_1 and the number of photons q in a laser are described by the well-known four-level rate equations [1]

$$\frac{dN_1(t)}{dt} = P(t) - \frac{\beta q(t) N_1(t)}{\tau} - \frac{N_1(t)}{\tau}, \tag{1a}$$

$$\frac{dq(t)}{dt} = -\frac{q(t)}{\tau_c} + \frac{\beta N_1(t)}{\tau}\left[q(t)+1\right], \tag{1b}$$

where P is the pumping fluence that is absorbed by the molecules inside the cavity, τ_c the cavity decay time, τ the spontaneous-emission life time, and β the beta factor of the laser. The beta factor is defined as the amount of spontaneous emission that contributes to the lasing mode, and can also be determined for a random laser [12].

From these rate equations Woerdman and coauthors have derived an equation for the frequency of the relaxation oscillations explicitly including the spontaneous emission:[13]

$$\omega_{\rm res} = \sqrt{\left(\frac{M-1}{\tau_c \tau}\right) - \frac{1}{4}\left[\frac{M}{\tau} - \frac{\beta}{\tau_c(M-1)}\right]^2}, \tag{2}$$

where $\omega_{\rm res}$ is the relaxation oscillations frequency, and M the scaled pump fluence, defined as the ratio of the absorbed fluence P and the threshold fluence P_{th}.

We apply this model to the multi-mode pulsed random laser by simply changing β to $\beta_{\rm mm}$ and τ_c to $\tau_{\rm c,mm}$, i.e. we use a mean cavity decay time and a mean beta factor to describe our multi-mode random laser. This simplification accurately describes the threshold behavior of random lasers [14].

III. EXPERIMENTAL APPARATUS

The random laser used in our experiments consists of a suspension of TiO_2 particles (mean diameter of 180 nm, volume fraction of 10%) in a solution of Sulforhodamine B in methanol (1 mmol/liter; pump absorption length, 104 μm; minimal gain length, 83 μm [15]). The suspension is contained in a fused silica capillary tube, with internal dimensions $100 \times 2 \times 2$ mm^3. To measure the mean free path of light in this sample, we performed an enhanced-backscatter cone experiment [16] and an escape function experiment [17]. We found a transport mean free path of 0.46 ± 0.1 μm at 633 nm (effective refractive index, 1.48 ± 0.04).

The samples were excited by a pump pulse at 532 nm, provided by an optical parametric oscillator (OPO) pumped by a Q-switched Nd:YAG laser (Coherent Infinity 40-100/XPO). The pump pulse had a duration of 3 ns and a repetition rate of 50 Hz. The pump light was focused with a microscope objective (water-immersed, numerical aperture NA, 1.2) onto the sample

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(focus area, $12 \pm 6\ \mu\mathrm{m}^2$), reaching an intensity in the order of 1 mJ/mm^2. The central wavelength of the emitted light is 594 nm [14], and the narrowing factor (defined as the spectral width of the emitted light far above threshold divided by the spectral width far below threshold) is 8. The light emitted by the random laser was collected by the same microscope objective. The emitted light was detected by a 25 GHz photodiode (New Focus 1404), read out by an oscilloscope (Tektronix TDS 7404, analog bandwidth, 4 GHz). The resulting time resolution was 100 ps. To obtain a good signal-to-noise ratio, the data shown is an average of 100 oversampled time traces. The pump light was filtered out of the detection path by use of a colored glass filter with an optical density of more than 4 at the wavelength of the pump laser.

IV. MEASURED RELAXATION OSCILLATIONS

The normalized time trace of the pump pulse and the normalized time trace of the total emitted light from the random laser far above threshold are shown in Fig. 1. Overall, the duration of the pump laser pulse is longer than the duration of the pulse of light the random laser emits. We see in the pulse emitted by the random laser first a fast decay, followed by a slower exponential decay. The fast decay is due to the stimulated emission in the random laser. In the second part of the decay the population inversion is no longer present, and the spontaneous emission causes a slower decay of intensity. These observations are in agreement with other random laser experiments.[8] In Fig. 1 relaxation oscillations in the emitted light are clearly visible near the peak intensity.

We measured the time evolution for different input fluences. In Fig. 2 the normalized intensity is plotted versus time for four different pump fluences [18]. The time traces are shifted vertically with respect to each other for clarity. The time trace at a pump fluence of 0.06 mJ/mm^2 is below threshold, while the time traces with higher pump fluences are above threshold. We observe that relaxation oscillations occur above threshold and become more pronounced when the pump fluence increases.

The frequency of the relaxation oscillations are computed from the time traces. We determine the times at which the intensity is at a local maximum. The difference of two consecutive local maxima Δt is the period, and the frequency of the relaxation oscillation ν_{rel} is given by $1/(\Delta t)$.

V. COMPARISON OF THE MEASUREMENTS WITH THE MODEL

We have inferred the relaxation oscillation frequency for different pump fluences from our measurements. In

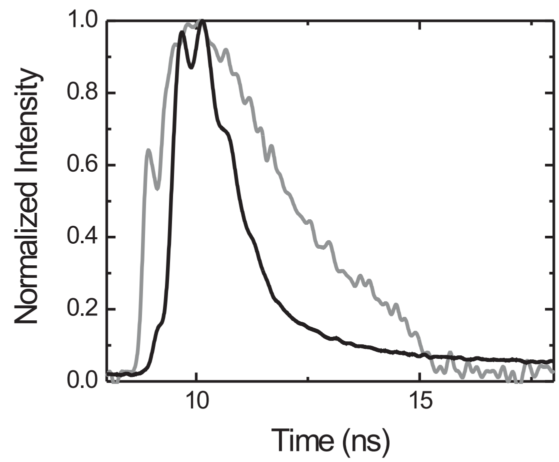

FIG. 1: Measured time traces of the pump pulse (gray) and emission output above threshold (black, input fluence $= 0.47$ mJ/mm^2) of a titania random laser. The pump pulse duration is much longer than the duration of the emitted light of the random laser above threshold. Relaxation oscillations in the emitted light are clearly visible near the peak intensity. The decay time of the emitted light is first dominated by stimulated emission. In the second part of the decay-curve, the spontaneous emission is dominating.

Fig. 3 the measured relaxation oscillations frequencies are plotted versus the scaled pump fluence M. The relaxation-oscillation frequency significantly decreases when the scaled pump fluence increases from 1 to 2. A further increase of the scaled pump fluence does not significantly change the frequency of the relaxation oscillations. The result of Eq. (2) is depicted for different cavity decay times. This cavity decay time is the only parameter that could not be directly determined by our experiment. The trend of the model for a fixed cavity decay time is that, in contrast to our measurements, the relaxation-oscillations frequency increases for increasing pump fluence. Only for large (> 3) normalized pump fluence the fit of the model for a cavity decay time of 5 ps fits reasonably.

The difference in the observed trend of the relaxation-oscillation frequency between the simple model and our experiments, is probably due to the size of the gain volume: The gain inside the gain volume of the random laser saturates at threshold [14]. A further increase of the pump fluence will lead immediately to a larger gain volume, and a corresponding longer cavity decay time. Apparently, this behavior saturates above a scaled pump fluence of 3.

We have used the distribution of the phase-delay time to determine the mean cavity decay time $\overline{\tau_c}$, given by [14]

$$\overline{\tau_c} = \frac{1}{8}\frac{L^2}{D}, \tag{3}$$

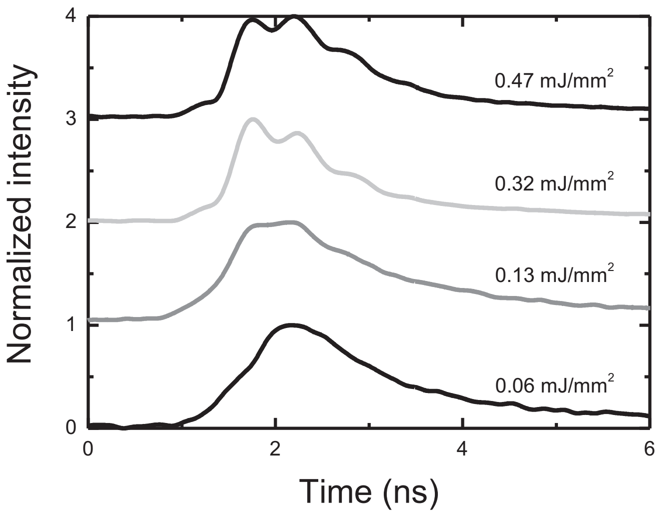

FIG. 2: Measured time traces of the emission intensities of a titania random laser for four different pump fluences. The traces are vertically shifted with respect to each other for clarity. Relaxation oscillations become more pronounced at higher pump fluences.

with L the length of the gain volume and, for non-resonant scattering, the diffusion constant $D = c_0\ell/(3n')$, where c_0 is the speed of light in vacuum, ℓ the transport mean free path and n' is the real part of the effective refractive index of the medium. For our titania random laser we find a $\overline{\tau_c}$ in the order of 10^{-13} s, in contrast to 10^{-12} s that is suggested by the agreement in Fig. 3 at $M = 3$. The difference between the two cavity decay times is a factor 10. This deviation could originate in part from the difference between the mean cavity decay time $\overline{\tau_c}$ of all modes and the mean cavity decay time of the lasing modes: Our experiment suggests that random lasing preferentially takes place in modes with a much longer than average cavity decay time.

VI. CONCLUSIONS

We have seen relaxation oscillations in our random laser, while looking at the time evolution of the total emitted light for different realizations of the sample. Multiple modes contributed to these time traces, and we averaged the time traces over several realizations of disorder of our random laser sample. The resulting time trace still showed relaxation oscillations: a weighted average of the oscillations of all the underlying modes.

The measured relaxation oscillations were compared with a simple model, based on a single-mode continuous-wave laser system. The observed trend of our experiments differs from the expected trend, due to the increase of the gain volume and the corresponding cavity decay time for an increase of the scaled pump fluence. The cavity decay time determined with the fit from the simple model is a factor 10 higher than the mean cavity decay time of our sample. Our experiment suggests that the modes contributing to random laser emission have a cavity decay time much longer than the average cavity decay time.

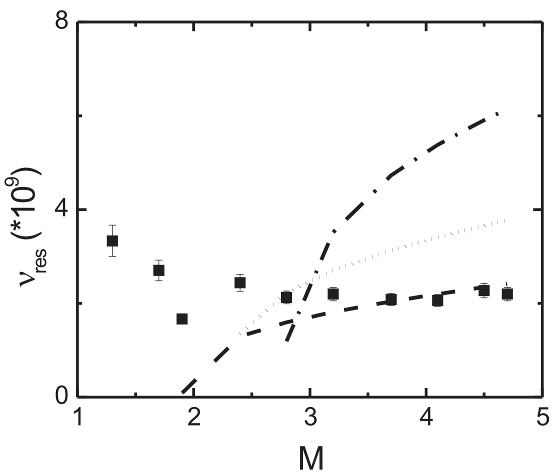

FIG. 3: Measured relaxation oscillations frequency as a function of the normalized pump fluence (squares). The simple model [Eq. (2)] is plotted for different cavity decay times: 0.7 (dash-dotted line), 2 ps (dotted line), and 5 ps (dashed line). The fit of the model for a cavity decay time of 5 ps fits reasonably for high pump fluences, but corresponds to a surprisingly large value of τ.

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